

Degradation of Glass Fiber Reinforced Polymer (GFRP) Material Exposed to Tropical Atmospheric Condition

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Abstract. Fibre reinforced polymers (FRPs) have emerged as popular materials for structural application in recent decades due to numerous of advantages. Despite the growing body of research on the use of glass fibre reinforced polymers (GFRP) composites in repairing and retrofitting the important structures such as oil and gas pipelines, the lack of comprehensive data on the long-term degradation mechanism for these materials is still impeding their widespread use in open-air structures repairs particularly in tropical climate locations such as Malaysia. Therefore, this paper presents an experimental investigation to determine the influence of tropical atmospheric condition on tensile properties of the GFRP. In this study, a set of GFRP samples were fabricated using epoxy resin as polymer matrix and woven E-glass fibres as reinforcing materials. These samples were exposed to tropical atmospheric condition in Malaysia for a period of four months. Tensile test was carried out for each sample before and after four-months period of exposure. The experimental tensile test results recorded a 15% reduction in tensile strength after 4 months of exposure as compared to its original strength. Further, the dominant failure mode of the exposed sample was characterized with longitudinal splitting of the fibres without completely breaking out. Overall, the tropical atmospheric condition has a noticeable impact on the GFRPs tensile strength degradations over the exposure duration.

Introduction

Fibre reinforced polymeric (FRP) materials have undergone a rapid growth replacing the use of the conventional metal materials in high-performance engineering applications mainly in aerospace, marine and automotive industries. The structure of FRP materials are typically comprised of a polymer matrix (e.g., epoxy, vinyl ester, or polyester) reinforced by fibres (i.e.: carbon, glass, aramid) to create a combination of the desired mechanical properties. The increasing in the applications of FRPs in the different industries is because of some noticeable advantages such as a high strength with low density, corrosion resistance, resistance to impact and friction, thermal stability, sound insulation, ease of handling and outstanding optical and electrical properties [1–5] Glass Fibre Reinforced Polymers (GFRPs) have been recently implemented in some engineering applications that involve repairing, rehabilitations or replacing the deteriorated existing structures. In marine and underground structures such as oil and gas pipelines, GFRP composites are an effective technique to retrofit any possible damages that might occur in those structures [6–9]

Atmospheric environment is defined as the air envelope around the Earth including its interfaces and interactions with the solid or liquid surface of the Earth. In other word, anything either living or non-living things on Earth in any situation that cover in air conclude as relate with atmospheric environment. Weather, moisture content, temperature, soil properties, salinity, humidity, wind direction and some parameters taken into consideration when discussing environmental factors in term of atmospheric environment. The application of GPRP composites in outdoor service (such as atmospheric environment) might be exposed to climate quantities such as solar radiation, heat, humidity, and rain. These climatic quantities would be more severe in tropical locations such Malaysia. In fact, these materials are more vulnerable to moisture and heat impact when function in such fluctuating conditions. The moisture in the humid climate is one of the main causes of the FRPs

degradation as it may penetrate into the matrix resulting in swelling, cracking, plasticization and fiber/matrix interfacial debonding [10–14]. Further, the moisture absorption by the fibrous composites increases with higher temperatures that characterizes their interfacial degradation rate [15]. Moreover, ultraviolet (UV) radiation is seen as a common degradation factor in the outdoor environment that mainly impacts the GFRP laminates with thinner thickness [13]. The UV radiation absorbed by the polymer causing a photo-oxidation reaction which mostly affects the surface of the composites [16]. Therefore, the GFRP composites in open-air applications might be exposed to multiple weathering factors leading to synergistic degradation processes.

Previous studies [10,13,17,18] have related the reduction in the mechanical properties of the GFRP composites (e.g., tensile strength, Young's Modulus, strain at failure) to the types of the environmental exposures. Nevertheless, this established correlation was based on artificial laboratory testing environments with accelerated aging exposure. Hence, this might not truly replicate the real situation of material degradation in the open-air. On the other hand, limited studies have considered the effects of the actual field conditions on degradation mechanism of GFRP composites such as seasonal conditions in Switzerland [19], urban environment in Portugal [20], hot weather in Saudi Arabia [21] and semi-arid climate condition in Kelowna, Canada [22]. Keller et al. [19] concluded that the eight years of winter exposure of a temporary GFRP truss bridge in Switzerland had caused tensile strength reduction up to 18 % whereas the Young's Modulus losses were negligible. Furthermore, the GFRP with unsaturated polyester or vinylester profile were exposed to natural environment condition in the urban region of Lisbon city centre in Portugal for 42 months had caused a degradation of 22 % in flexural strength, and of 33% and 37% in tensile and flexural moduli, respectively [20]. In addition, degradation of the tensile properties for GFRPs bars in two field conditions of the Saudi Arabia was 2% which is small reduction in hot weather that indicates that the hot weather in the middle east region has less effect on GFRP composites [21]. Lastly, a semi-arid climate with dry, sunny summers, cold, cloudy winters and all four seasons in Kelowna, Canada was a field environment condition to investigate the effect of 9 months exposure on the GFRPs degradation and it was found that the effect level on mechanical and physical properties of GFRPs due to fiber orientation, cure condition, and gel coating can be altered over time under natural weathering conditions [22]. Therefore, the abovementioned studies confirmed that the different field environmental conditions in different geographical locations around the world can lead to completely different degradations of the FRPs mechanical properties.

The susceptibility of GFRP durability attributed to individual or synergistic impact of humidity, ultraviolet, heat and rain in tropical climatic locations is a significant issue that should be examined. Thus, this study provides understanding on the natural long-term degradation of glass fibre-reinforced polymer under tropical atmospheric condition in Malaysia. This study focused on the evaluation of tensile strength as a measure to assess the degradation.

Methodology

Samples preparation

In this study, the FRP samples were prepared using conventional wet-layup method as stated per supplier's guidelines. Further, the proportion of epoxy and hardener in matrix was based on the recommended mixing ratio by the supplier. The materials used in preparing of the GFRP samples were E-glass fibres that contains more than 60 % of silica, liquid epoxy resin and liquid hardener as curing agent. Moreover, due to the commercial confidentiality, the mechanical properties of "stand alone" material are not provided. Basically, the composite laminate of 300mm x 250mm x 3mm was prepared from four layers of 0°/90° bi-directional woven E-glass fibre mats, epoxy resin and hardener (curing agent) using hand layup fabrication technique. Each layer of woven E-glass fibre mat was cut precisely into the above-mentioned dimensions as shown in Fig. 1(a). During the hand layup preparation process, the first layer of E-glass fibre mat was placed by hand on baking paper and then a mixture of 24.15g of epoxy resin and 10.85g of hardener was poured thoroughly on the placed reinforcing fibre layer (see Fig. 1(b)). After that, a roller was used to consolidate the layer ensuring an entirely wetting of fibre mats and removing the entrapped air and voids in the matrix as shown in Fig.

1(c). Then the same process was applied for the subsequent three layers forming the final composite product. Lastly, the 300mm x 250mm composite laminate was left for a while to be partially cured and then cut into twelve equal pieces with 25mm width and 250mm length as seen in Fig. 1(d). The cut edges of each sample were sealed with epoxy resin to stop instantaneous ingress of moisture into the composites when they are exposed to the open-air environment.

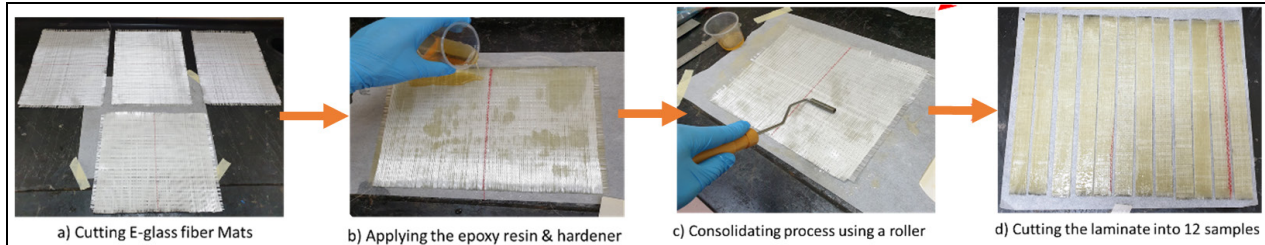


Fig. 1. Preparation process of GFRP samples using hand layup method.

Exposure condition

In this stage, a total number of 11 samples were prepared for glass fibre-reinforced polymers (GFRPs). Six (6) out of the eleven (11) samples were used as reference (control sample) without any exposure. On the other hand, the remaining five (5) samples were exposed to tropical atmospheric condition in Malaysia for a period of four (4) months to evaluate the impact of the climatic quantises (e.g., heat, solar radiation, humidity and rain) on the samples' durability. Fig. 2 shows the GFRPs sample during the exposure.



Fig. 2. GFRP samples exposure.

Tensile test

Eleven GFRP specimens were tested under a direct tension load using universal testing machine (UTM) with 50 KN capacity. Five samples were exposed to tropical atmospheric condition as described in previous section and the other six samples were used as reference without any exposure. The direct tension test was performed by employing a displacement control method with displacement rate of 2mm/min based on ASTM D3039 [23]. A strain gauge was attached at the middle part of each sample to measure the strain value. The applied displacement, the load response and the strain response were all recorded during the test time. The experimental tensile test set up are shown in Fig. 3. The overall details of specimens including the exposure environment, exposure duration, number of samples, geometry details, test employed and loading protocol and rate are summarized in Table 1.

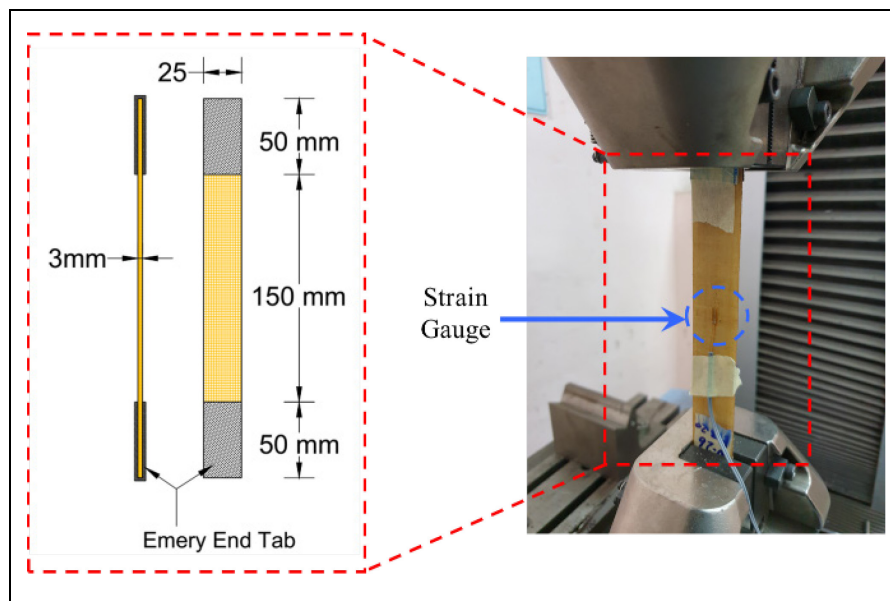


Fig. 3. Experimental tensile test set up.

Table 1. Summary of samples details.

| Samples Type | Control Samples | Exposed Samples |
|------------------------|-----------------------|------------------|
| No. of Samples | 6 | 5 |
| Dimensions (L x W x t) | 251.3x25x3.43 mm | 252x25.3x3.43 mm |
| Exposure duration | - | 4 months |
| Test used | Uniaxial tensile test | |
| Standard | ASTM D3039 [23] | |
| Loading protocol | Displacement control | |
| Loading rate | 2mm/min | |

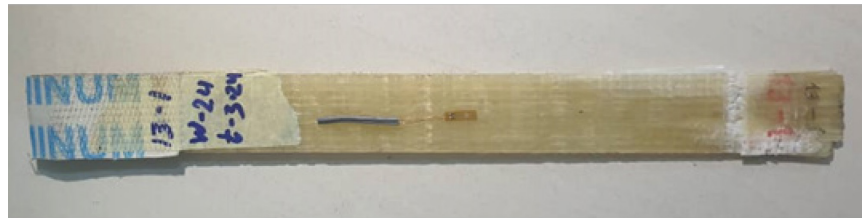
Results and Discussion

Mode of failures

The dominant mode of failure in the control samples occurred near the top emery end tab with brush-like failure characterized by the long splitting of the fibres as shown in Fig. 4(a). This failure mode typically takes place in unaged or damaged material [24]. The transverse failure with clean cut of both matrix and woven glass fibre mats was also noticed in two samples (see Fig. 4(b)).



a) Brush-like Failure with longitudinal splitting



b) Transverse fracture failure

Fig. 4. Modes of failure for control samples.

In the atmospheric exposure samples, the tensile failure for four samples were occurred within the gauge length as multiple interlaminar cracks took place in the same direction of the longitudinal fibres causing splitting failure without completely fracture of these fibres as seen in Fig. 5(a). This failure might be caused due to the possibility of the water penetration during the exposure period that weakens the fibre/matrix interface bond and with the less resistance of the transvers fibres that can eventually result in an inadequacy in the integral load transfer capacity. On the other hand, transverse failure mode with clean cut of the fibres was observed in one sample as shown in Fig. 5(b).



a) Longitudinal splitting of fibers with partial breaking out



b) Transverse fracture of the laminate

Fig. 5. Failure modes for 4 months exposed samples.

Surface colour changes

The surface colour of the samples in this study changed from yellow and shiny to a noticeably brown after the 4 months exposure as shown in Fig. 5. The same appearance changes was also noticed in some studies exposed field conditions [20,22]. Sousa et al. [20] observed that significant changes occurred in the gloss and colour of the exposed samples. Similarly, Heinrick et al [22] noticed that after 9 months of exposure, the samples endured some physical changes involving discoloration, cloudy appearance within the resin matrix. Both studies have linked the surface colour changes to the impact of UV radiation. In further explanation, the photodegradation of polymer matrix is initiated by interaction between the UV photons and polymer molecule chains resulting in surface discoloration, surface oxidization and bond disassociation [25,26]. Furthermore, UV impact is more intensified when compounded by other climatic quantities such as temperature and moisture [27]. Therefore, the longer exposure to these climatic quantities, the more colour and lustre of GFRP surfaces will be changed.

Tensile properties results

The results of tensile properties for control and atmospheric exposure samples are summarized in Table 2 including ultimate strength, modulus of elasticity and strain at failure for each sample in controlled and exposed condition. Further, the average magnitude for the above-mentioned properties was determined along with calculated standard deviation and coefficient of variation for each as given in summary portion in Table 2. The standard deviation is preceded by a plus-minus symbol (\pm) indicating the data variability around the average value of the entire samples. The tensile strength for control or exposure samples was taken as the average value of their respective ultimate tensile strengths. Further, the Young's modulus was calculated as slope of stress-strain curve on the incremental basis.

Table 2. Tensile properties of control and exposure GFRP material

| Sample Type/Label | | Ultimate Tensile Strength (Map) | Young's Modulus, E (GPa) | Strain at Failure (mm/mm) |
|-------------------|-------------------------|---------------------------------|--------------------------|---------------------------|
| Control Samples | CS1 | 307.39 | 23.37 | 0.00920 |
| | CS2 | 304.76 | 18.60 | 0.01191 |
| | CS3 | 297.80 | 20.84 | 0.00999 |
| | CS4 | 328.63 | 20.63 | 0.01183 |
| | CS5 | 271.50 | 19.88 | 0.00892 |
| | CS6 | 335.51 | 20.12 | 0.01114 |
| | Average | 307.60 | 20.57 | 0.01050 |
| | Standard Deviation | 22.93 | 1.58 | 0.00131 |
| | Coefficient of Variance | 7.45 | 7.68 | 12.48 |
| Exposed samples | ES1 | 253.96 | 19.88 | 0.00868 |
| | ES2 | 249.14 | 24.39 | 0.00767 |
| | ES3 | 300.86 | 23.69 | 0.00944 |
| | ES4 | 245.03 | 19.40 | 0.00899 |
| | ES5 | 262.37 | 19.53 | 0.01029 |
| | Average | 262.27 | 21.38 | 0.00901 |
| | Standard Deviation | 22.51 | 2.45 | 0.00097 |
| | Coefficient of Variance | 8.58 | 11.46 | 10.77 |
| Summary | Control Samples | 307.60 \pm 22.93 | 20.57 \pm 1.58 | 0.01050 \pm 0.00131 |
| | Exposed Samples | 262.3 \pm 22.51 | 21.38 \pm 2.45 | 0.00901 \pm 0.00131 |
| | Reduction (%) | 15% | - 4% | 14% |

The stress-strain curve for the control and four months atmospheric exposure samples are shown in Fig. 6. Basically, both stress-strain curves exhibited almost linear behaviour until the failure which indicates the GFRP composites are brittle in their behaviour. Furthermore, the impact of the atmospheric exposure on the tensile strength behaviour of the GFRP samples in four-month period can be seen clearly in their stress-strain curve which slightly displays a lower tensile stress than control samples until the failure.

Based on Table 3, the ultimate tensile strength of GFRPs samples in control condition was recorded with value of 307.6 MPa. However, with four months exposure in the atmospheric tropical condition, the tensile strength has been reduced by 15% reaching to 262 MPa. This reduction in strength can be related to the changes occurred in the fibre/matrix interface because of the synergistic damage by the moisture penetration and the temperature fluctuations. On the other hand, the modulus of elasticity for four-month exposure samples were recorded with the value of 21.38 GPa which is 4% higher than the one recorded for control samples. The reason according to Mourad et al.[28] is related to the

possible swelling in the samples during the exposure that leads to increase in the modulus and lower in strain at failure. Furthermore, the strain at failure for atmospheric exposed samples was recorded with value of 0.00901 which is 14 % lower than samples tested in control condition.

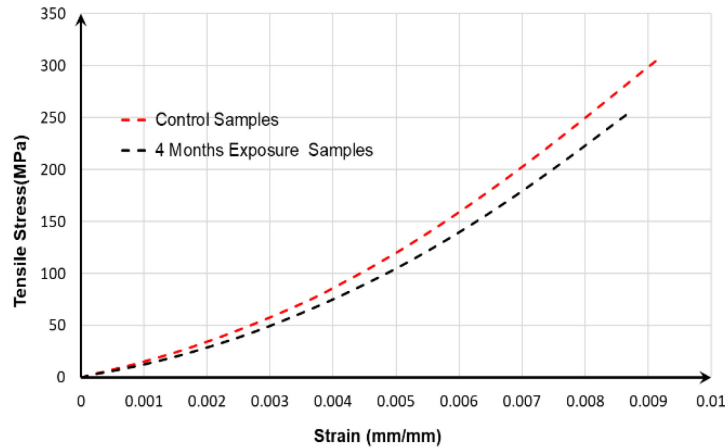


Fig. 6. Stress-strain curves for control and 4 months atmospheric exposure samples

To better understand the effect of the actual field environmental conditions on the tensile properties of GFRP, the present study was compared with some of the reported studies in the literature on the basis of the FRP type, in-situ environment, maximum duration of exposure, tensile strength reduction and modulus of elasticity reduction as shown in Table 3. The maximum reduction of the tensile strength of GFRP composite was 18% under effect of seasonal condition (winter) in Switzerland however, this reduction occurred after 8.5 years of exposure. Furthermore, the tensile strength reduction of GFRP composite after 3.5 years of exposure to urban environment in Lisbon, Portugal was 14%. Therefore, in the present study, 15% reduction of the GFRP tensile strength within only 4 months of exposure indicates how the significant impact of tropical atmospheric condition in Malaysia on the GFRP degradation. On the other hand, significant loss of modulus of elasticity was only noticed for the GFRPs exposed to the natural aging urban environment in Lisbon, Portugal with 33% of Young's Modulus reduction for E-glass/ polyester GFRP pultruded profile and 24% reduction for E-glass /vinylester GFRP pultruded profile. In contrary, no significant loss of modulus of elasticity was noticed for GFRP composite in seasonal condition in Switzerland (winter) and in hot weather in Saudi Arabia. Overall and based on the comparison being made that the different field environments on different locations around the globe can induce dissimilar degradation of the FRP tensile properties.

Table 3. Comparison between the tensile test result of the present study and similar research

| Reference | FRP type | In-situ environment | Maximum duration | Tensile strength reduction (%) | Young's Modulus reduction (%) |
|---------------|---|---|-----------------------|--------------------------------|-------------------------------|
| Present study | E-glass /epoxy GFRP coupons | Tropical atmospheric condition in Malaysia | 4 months | 15% | -4% |
| [19] | GFRP isophthalic polyester pultruded coupons (taken from truss pedestrian bridge) | Seasonal conditions in Switzerland (winter) | 8.5 years | 18 % | No significant loss |
| [20] | E-glass/ polyester GFRP pultruded profile | Urban environment in Lisbon, Portugal | 42 months (3.5 years) | 14% | 33% |
| | E-glass /vinylester GFRP pultruded profile | | | 3% | 24% |
| [21] | E-glass/vinylester GFRP bars | Hot weather in Saudi Arabia | 18 months | 1-2% | No significant loss |

Conclusion

In this study, glass fibre-reinforced polymers (GFRPs) were exposed to a tropical atmospheric condition in Malaysia for a period of four months to understand their degradation mechanism in the longevity term. By exposure to climatic quantities in tropical locations (e.g., ultraviolet radiation, moisture, temperature fluctuations, humidity, and rain), the tensile properties of GFRPs are vulnerable to degradation over time. Based on the tensile test findings for the GFRPs under four months of exposure in tropical atmospheric conditions, the following conclusions are drawn:

- Fibre splitting and fibre/matrix fracture with clean cut were the dominant failure modes after 4 months of exposures.
- The GFRPs surface colour changed from yellow and shiny to a noticeably brown over the exposure period.
- Tensile test results of GFRPs recorded a 15% reduction in tensile strength after 4 months of exposure as compared to its original strength.
- The modulus of elasticity for four-month exposed samples was 4% higher than control samples.
- Degradation of GFRP composites varied according to environment type being exposed and the location of that environment.

Therefore, obtained findings in this study can help the engineers in the oil and gas industry to consider the degradation effect of the tropical atmospheric condition on the FRP composites application in repairing and retrofitting the deteriorated pipelines.

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